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Naval Surface Warfare Center
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Feasibility of Using Artificial Neural Networks With Electrochemical Impedance Spectroscopy Data From Coated Steel

by
Harvey P. Hack and M. Allen Matteson

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ABSTRACT

Electrochemical impedance spectroscopy (E.I.S.) techniques can provide information about the condition of protective coatings on steel marine structures. Currently, an expert is required to interpret the data produced from an E.I.S.. measurement, classifying the coating as "good" or "poor" or identifying the data as "bad." This limits the use of E.I.S.. techniques to experienced operators. If the E.I.S.. technique is to be used for production by inexperienced operators, measurements must be classified automatically.

This investigation uses artificial neural networks (ANN) to develop an automated E.I.S. data classifier. ANNs were trained with a large database of measurements on known good or poor coatings, including some bad data. The ANNs were tested with E.I.S.. data not included in the training set. A variety of measurement signal processing schemes and network structures was evaluated. ANNs were developed which can accurately determine if the coating is good or poor and whether measurement problems produced bad data.

CONTENTS

	Page
Abstract	iii
Administrative Information	vi
Acknowledgments	vi
Abbreviations	vi
Introduction	1
Background	2
E.I.S. for Coatings on Steel	2
Artificial Neural Networks	2
Procedure	6
E.I.S. Data Source and Treatment	6
Neural Network Setup	6
Results and Discussion	10
Conclusions	22
References	23
Initial Distribution	25
Standard Form 298	27

FIGURES

1. Impedance graphical analysis.	3
2. Biological neuron.	4
3. Artificial neuron network processing element.	4
4. Simple artificial neural network structure.	5
5. Network 6—"good" coating prediction.	11
6. Network 6—"poor" coating prediction.	12
7. Network 6—"bad data" prediction.	13
8. Network 17—"good" coating prediction.	14
9. Network 17—"poor" coating prediction.	15
10. Network 17—"bad data" prediction.	16
11. Network 3—"bad data" prediction.	17
12. Network 4—"bad data" prediction.	18
13. Network 5—"bad data" prediction.	19

TABLES

1. Different input schemes. 8
2. Different output schemes. 8
3. Learning rule and parameters. 9
4. Networks studied. 20
5. Results. 21

ADMINISTRATIVE INFORMATION

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ABBREVIATIONS

ANN	Artificial neural networks
CARDEROCKDIV, NSWC	Carderock Division, Naval Surface Warfare Center
E.I.S.	Electrochemical impedance spectroscopy
PAR	E.G. & G. Princeton Applied Research (measuring system)
PE	Processing element

INTRODUCTION

Electrochemical impedance spectroscopy (E.I.S.) is a measuring tool which has gained wide acceptance for its ability to determine the quality of organic coatings on steel in seawater and other aqueous media.[1] For this reason, its use has become fairly common in the laboratory for this application.

Each measurement is a series of data points, which are typically presented graphically and are analyzed by a variety of computational methods.[2] The calculations, although not computationally complex, can be difficult to understand for those not trained in the E.I.S. technique. Alternative analysis techniques involve working with the shape of the graph segments.[3] For this reason, data are usually analyzed by highly trained personnel. Even for them, the analysis process can be time consuming, so only a limited amount of data can be analyzed. Data interpretation and, therefore, conclusions about coating performance are sometimes dependent on the individual performing the analyses.

The Carderock Division, Naval Surface Warfare Center (CARDEROCKDIV, NSWC) has been using E.I.S. to evaluate organic coatings on steel for many years.[4–8] During this time, the technique has progressed from a laboratory curiosity to a reliable method for analyzing coating performance. Most recently, CARDEROCKDIV, NSWC has encouraged the development of portable devices for measuring very high impedances associated with these coatings.[6] Field measurements are now possible, but analysis of the data from these measurements is still difficult.

Coating performance is highly variable. Analysis of the performance of a coating system therefore requires many measurements on many samples to get a statistically valid result.[9] The high cost of performing analyses because of the large amount of time spent by highly trained individuals is a barrier to the wide use of E.I.S. for evaluating coating performance in the field. A technique for low-cost rapid analysis of E.I.S. data on coated steel is needed for the technique to gain widespread use, particularly for field evaluations.

Artificial neural network software is an ideal choice for pattern recognition and is, thus, worthwhile to explore as a technique to automate E.I.S. data analysis. Artificial neural network analysis is not necessarily dependent on the assumption of a specific electrical equivalent circuit to model the coating, like the more traditional techniques of analysis. The objective of this work was to determine the feasibility of using artificial neural networks to analyze E.I.S. data accurately from organic-coated steel. If the use of this computational technique was deemed possible, the next objective was to determine what type of network would work most efficiently to analyze coating data. Another follow-on objective not reported herein was to optimize the network architecture and the training data sets.

The ultimate aim of this work was to develop software which could be loaded into the computers that drive the portable E.I.S. apparatus and could give real-time analysis of the data from the E.I.S. measurements. Such a device—a combined measurement and analysis tool—could be used by technicians in a laboratory or could be taken to an industrial site such as a shipyard to perform field measurements and give immediate feedback about coating condition. Use by less skilled personnel would lower the cost of these measurements, thus promoting their use.

BACKGROUND

E.I.S. FOR COATINGS ON STEEL

An E.I.S. measurement is taken by applying a small sinusoidal voltage, typically no more than 10 or 20 mV relative to the rest potential of a coated steel surface, which is immersed in an electrolyte, and measuring the magnitude and phase shift of the resultant current.[10] From this, a complex impedance value for the coated surface can be calculated.[11] The measurement frequency is then changed and another data point taken. This process is repeated over a frequency range of typically a few tens of millihertz to 100 kHz in roughly logarithmic intervals to constitute one complete measurement set.

Data generated by this technique are usually displayed graphically, typically using Bode diagrams that plot either phase angle or the log of the magnitude of impedance or both against the log of frequency.[12] These graphs are then analyzed by determining features that relate to coating performance,[13] as illustrated in Figure 1. Level sections of the magnitude plot corresponding to areas of zero phase shift are interpreted as pure resistances—typically the polarization resistance, the coating pore resistance, the electrolyte resistance, or their sum. Sections of the magnitude plot at a –1 slope corresponding to areas of constant 45-degree phase shift are interpreted as capacitances—typically the coating capacitance, the interfacial pseudocapacitance, or their sum. Analysis is typically done either by curve fitting to an electrical equivalent circuit for the assumed coating behavior[13] or by manually determining values of impedance plateaus—break-point frequencies where transition from –1 slope to level behavior occurs—and the location and slope of certain segments of the curve. When analysis does not use curve fitting, calculations must then be made from the observed shapes and positions of curve segments to come up with values such as coating capacitance, coating resistance, double-layer capacitance, polarization resistance, and diffusional impedances. These parameter values are then used to determine whether the coating integrity is good or poor.

ARTIFICIAL NEURAL NETWORKS

Artificial neural networks (ANNs) simulate in an extremely simple manner the workings of the human brain.[14] The basic cellular computing unit of the brain is the neuron. Human brains are composed of billions of interconnected neurons, which communicate through the release of chemical neurotransmitters that can cause or inhibit electrical signals within a neuron. Each neuron receives input from many different neurons and in turn produces outputs to many other neurons. In the process of learning, the ability of an active neuron to affect other neurons is modified by affecting the strength of its connection to the other neurons. A simplified diagram of a biological neuron is shown in Figure 2.

The artificial analogy to the biological neuron is called a processing element (PE), represented by the diagram in Figure 3. Like their biological analogues, each PE receives inputs from many PEs and sends outputs to many other PEs. The inputs are usually weighted, combined by simple summation, then analyzed by a transfer function in the PE. Often, the transfer function is simply a threshold value, causing the PE to pass output only if the weighted, summed inputs exceed a certain value.

Processing elements are organized into layers in ANNs, as shown in Figure 4. Usually there is an input buffer layer, an output buffer layer, and, optionally, one or more

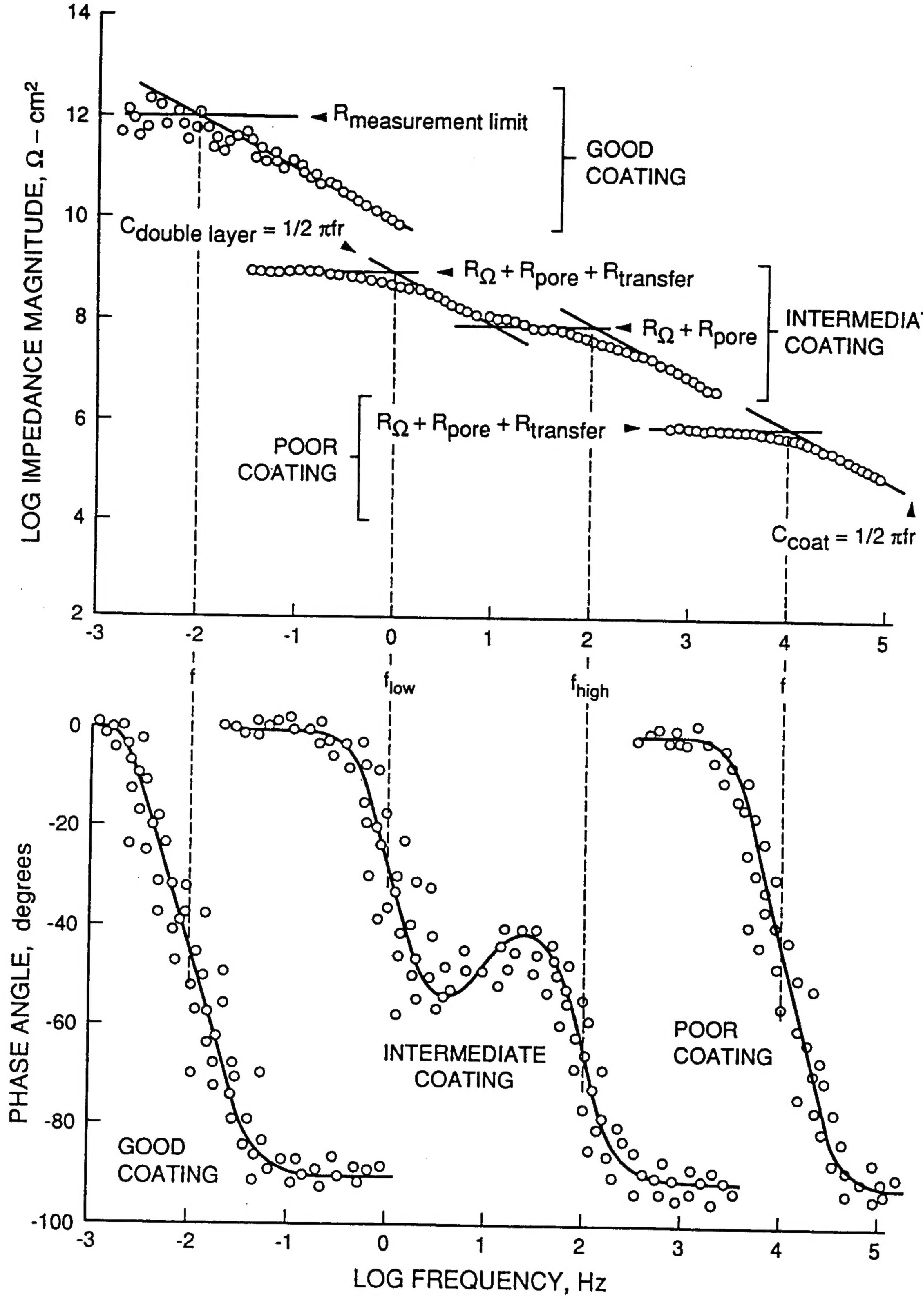

Figure 1. Impedance graphical analysis.

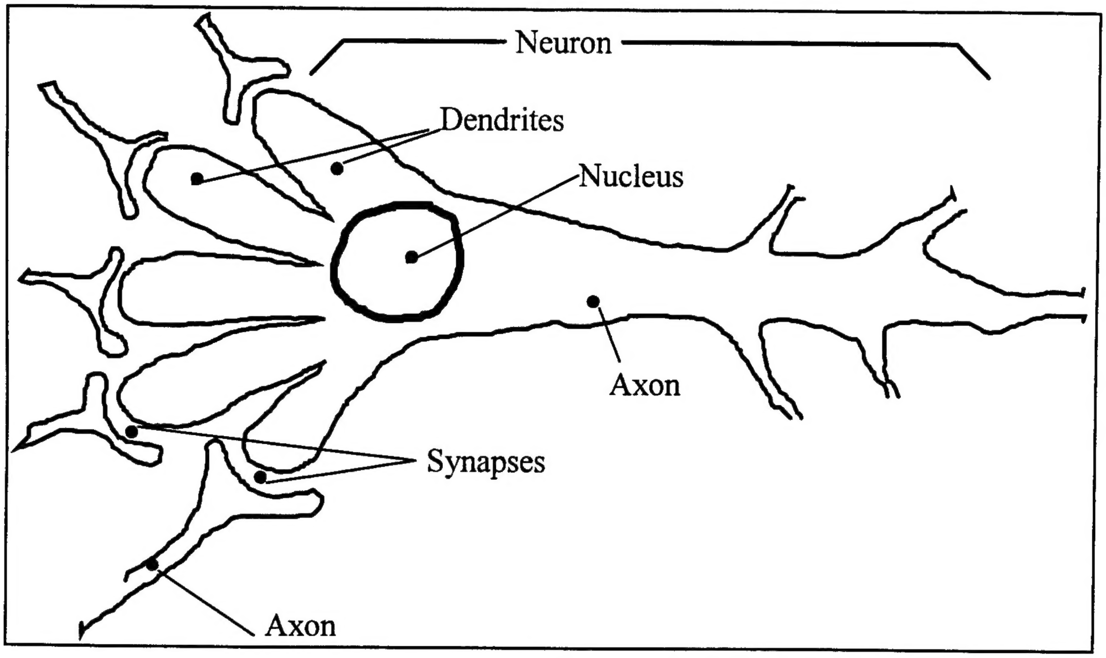

Figure 2. Biological neuron.

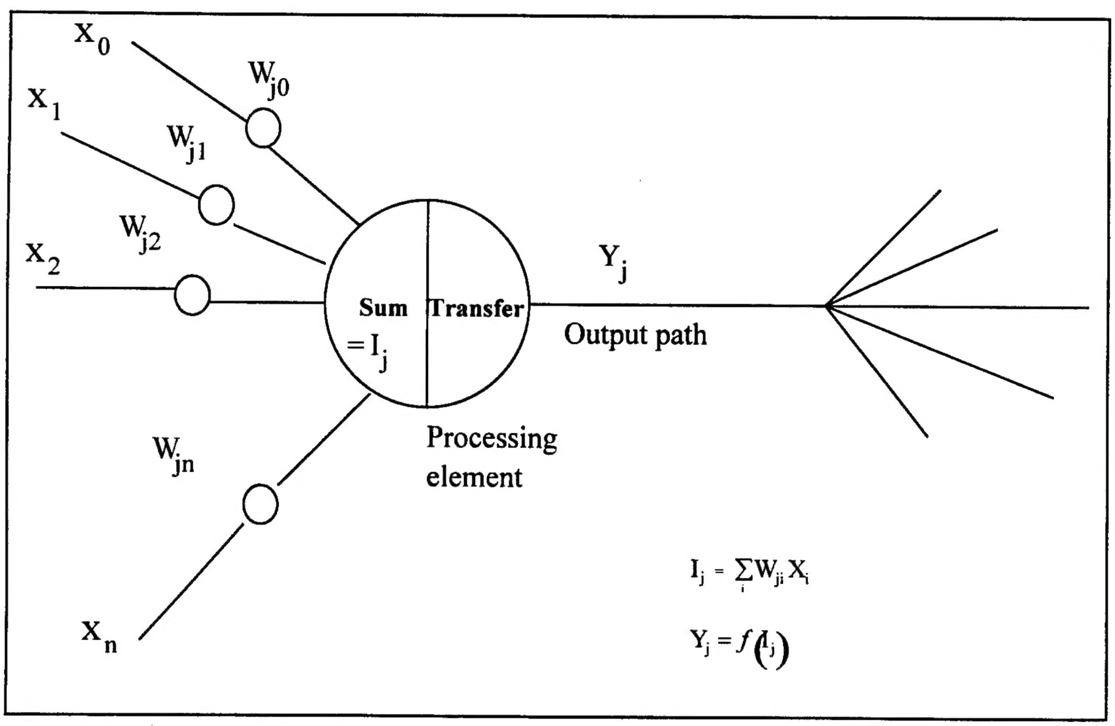

Figure 3. Artificial neuron network processing element.

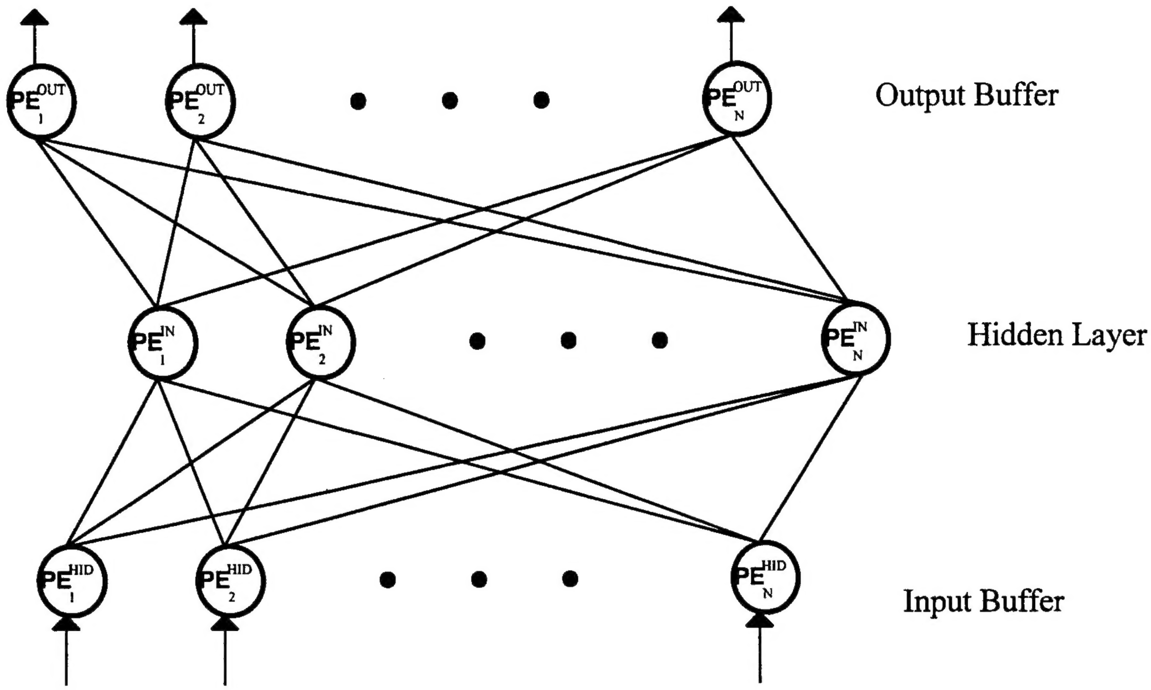

Figure 4. Simple artificial neural network structure.

layers in between, called hidden layers. In a typical ANN, each PE of each layer is connected fully with each PE in the layers directly before and after it. ANNs learn to associate certain inputs with certain outputs by assigning weights to the connections between two PEs. Learning is achieved through a learning rule which adapts or changes the connection weights of the network in response to the example inputs and, optionally, the desired outputs for those inputs. In supervised learning, for each input stimulus, a desired output is presented to the system and the network gradually configures itself to achieve that desired stimulus/output mapping. The connection weights are the memory units of a neural network. The values of the weights represent the current state of knowledge of the network. All of the features and functions of a typical back propagation ANN can be readily represented or modeled in computer code.

Certain neural computing memories are associative, i.e., if the trained network is presented with a partial input the network will choose the closest match in memory to that input and generate an output which corresponds to a full input. The nature of neural network memory leads to reasonable network response even when presented with incomplete, noisy, or previously unseen input.

The advantages of ANNs are that they are able to classify data without firm rules and they are more suited to problems that are ill-defined or that would require a large number of rules.[15] Networks develop their own set of rules for a classification problem through the supervised learning process.[16] Once the network has configured itself for the given training set, it is capable of correctly classifying input values it has not experienced before, as long as the set of data used for training the network is representative of the system to be classified. The technique is intrinsically able to handle complex, noisy,

irrelevant, and partial information. A disadvantage of network techniques is that unlike traditional expert systems, it is not easy to obtain information about how the network reached a conclusion.[17] Because of this, it is important to have a high degree of confidence in data used to train the network. If the training data do not accurately represent the system, the resulting network will be of little value.

PROCEDURE

E.I.S. DATA SOURCE AND TREATMENT

As described in greater detail elsewhere,[18–20] mild steel panels were coated with various materials, principally epoxy-polyamide paints such as those used in naval construction, although latex and other coating materials were also used. Each panel was fitted with a cylindrical glass electrochemical cell and sealed against the panel with an O-ring and a spring clamp. Artificial ocean water* was used to fill the cell, and a carbon counter electrode and silver/silver-chloride reference electrode were fitted for the duration of each E.I.S. measurement. A series of measurements was taken for each panel periodically over several years exposure.

The measurements were taken with an E.G. & G. Princeton Applied Research (PAR) model 273 potentiostat and model 5208 lock-in analyzer using model 378 software. Each measurement was made as a single sine (frequency-by-frequency) run starting at 100,000 Hz and stepping down to roughly 5 Hz in logarithmic progression with seven frequencies per decade. This was followed by two white noise (Fourier transform) runs with base frequencies of 0.1 and 0.005 Hz. Data at each frequency consisted of a real and an imaginary component of impedance, normalized to ohms per centimeter squared.

NEURAL NETWORK SETUP

Each measurement was rated regarding whether that measurement was on a good coating or a poor coating and whether the data were bad (because of a variety of experimental problems that can occur when taking E.I.S. measurements). These ratings were based on visual observation of the coating and the shape of the impedance curves. The input to the network consisted of these ratings and the impedances. For some networks, values of various impedances, capacitances, and break-point frequencies were determined by manually fitting the impedance curves, as shown in Figure 1. These values were added to the input data set to train the network and to test the network's calculation of these intermediate values. Results of networks used to predict impedances, capacitances, and break-point frequencies were not complete as of this writing but will be presented in a later report.

The E.G. & G. PAR measuring system saves each measurement in a file in PAR format. Some data processing was necessary in order to convert the measurement data into a form appropriate for processing by a neural network. The following steps detail the processing used to generate the training and testing data files for the neural network study:

1. PAR measurement data files were first converted from the PAR format to ASCII format using software developed at CARDEROCKDIV, NSWC. This created a single file for each measurement. Each measurement con-

*Per ASTM D 1141, "Specification for Substitute Ocean Water."

sisted of 71 data pairs corresponding to the real and imaginary parts of the complex impedance value for each of the 71 discrete frequency values.

2. Each measurement was assigned a specification of "good coating," "poor coating," or "bad data" based on a traditional, i.e., manual, evaluation of the data point. There were 71 "poor coating" measurements, 70 "good coating" measurements, and 13 "bad data" measurements.
3. All the ASCII data files for "good coating" measurements were then combined into a single Excel spread-sheet file.
4. All the ASCII data files for "poor coating" measurements were combined into a second Excel spread-sheet file.
5. All the ASCII data files for "bad data" measurements were combined into a third Excel spread-sheet file.
6. Each measurement consisted of the following values in the order listed:
   a. An arbitrary identifying number;
   b. Seventy-one frequency pair values (real and imaginary part for a total of 141 numbers);
   c. A "good coating" value;
   d. A "poor coating" value;
   e. A "bad data" value.
7. A "training" data set was created by random selection of half of the "good coating" measurements, half of the "poor coating" measurements, and half of the "bad data" measurements and combining these into a single file.
8. A "test" data set was created by combining the remaining half of the measurements into another file.
9. For certain networks the input data were processed in order to determine the sensitivity of the network to the form of the impedance data. This processing involved calculating the natural log of the magnitude of the impedance value for each of the 71 frequency values. This log was determined as the square root of the sum of the squares of x_λ and y_λ, where x_λ is the real part of the impedance at frequency λ and y_λ is the imaginary part of the impedance at frequency λ. This reduced the number of values for each E.I.S. measurement by one half because each pair of impedances at a given frequency became a single impedance magnitude number.
10. For other networks, the data were manipulated in other ways to allow for variation in the input and output schemes selected. These schemes are described hereinafter in more detail. The input (stimulus) values for the networks were the impedance information, while the response (desired output) of the networks was the coating classifications, i.e., "good" coating, "poor" coating, or "bad" data.

Neural networks were defined, configured, trained and tested using development software called NeuralWorks.[16] This application provided a means of describing the network in terms of the number of input PEs, the number of hidden layers and the number of PEs in each, and the number of PEs in the output/response layer. Once that was accom-

plished, the user could then select the type of learning rule to be used and the training parameters to be used during the training phase. It was then necessary to specify the file which contained the data to be used for training the network and optionally the file which contained the data to be used for testing the network.

A large number of networks was defined, configured, and tested. Several variations of the form of the input data were used as well as tests of several different numbers of hidden layers and processing elements in each hidden layer. Three different output schemes were tested as well, but not for all of the input schemes. Table 1 shows the different input schemes that were used.

Table 1. Different input schemes.

A	Real and imaginary impedance value for each of the 71 frequency bands
B	Log of the magnitude of impedance values for each of the 71 frequency bands
C	Real and imaginary impedance value for only those frequency bands of 1 hz and below
D	Log of the magnitude of impedance values for only those frequency bands of 1 Hz and below
E	Real and imaginary impedance value for only those frequency bands of 10 Hz and above
F	Log of the magnitude of impedance values for only those frequency bands of 10 Hz and above

Table 2 shows the three different output schemes that were used.

Table 2. Different output schemes.

Output Scheme	Measurement Type	Output Value
	Good Coating	Good Coating Value=1.0
		Poor Coating Value=0.0
		Bad Data Value=0.0
	Poor Coating	Good Coating Value=0.0
A		Poor Coating Value=1.0
		Bad Data Value=0.0
	Bad Data	Good Coating Value=0.0
		Poor Coating Value=0.0
		Bad Data Value=1.0

Output Scheme	Measurement Type	Output Value
B	Good Coating	Good Coating Value=1.0
		Poor Coating Value=0.0
		Bad Data Value=0.0
	Poor Coating	Good Coating Value=0.0
		Poor Coating Value=1.0
		Bad Data Value=0.0
	Bad Data	Good Coating Value=0.5
		Poor Coating Value=0.5
		Bad Data Value=1.0
C	Good Coating	Good/Poor Coating Value=1.0
		Bad Data Value=0.0
	Poor Coating	Good/Poor Coating Value=0.0
		Bad Data Value=0.0
	Bad Data	Good/Poor Coating Value=0.5
		Bad Data Value=1.0

The procedure for evaluating each data input and output scheme combination was to first generate the training and test data files with the input and output scheme to be tested. Next, using NeuralWorks Professional II software, a network was specified with input processing elements equal to the number of input values in the measurement for the input scheme being tested. For the first test of an input/output scheme combination, the network configuration had no hidden layers. Finally, the number of output processing elements was specified to equal the number of output values in the output scheme being tested. The network training rule and training specifications were then determined and the network was ready to begin the training phase. For every network considered in this study the learning rule and training parameters were identical; the only differences between networks were the input scheme, the output scheme, and the number of hidden layers and processing elements in those hidden layers. For this study the learning rule and learning parameters (see Table 3) were as follows:

Table 3. Learning rule and parameters.

Learning Rule	Norm-Cumulative
Transfer Function	TanH
Momentum	0.4
First Transfer Point	10,000
Learning Coefficient Ratio	0.5
Training Iterations	100,000

The significance and effect of each of these parameters are described in detail elsewhere.[14] At the end of the training phase the network was tested by presenting it with the data contained in the test file which the network had not seen or used during the training phase. The results of the testing were automatically placed in a data file by the NeuralWorks Professional II software in a format that included the desired or actual values. The network was then saved and the next network to be tested configured. For a series of tests for a single input and output scheme, the next network configuration to be tested was one with a hidden layer. In every case, the number of processing elements in a hidden layer was approximately equal to one half the number of processing elements in the layer preceding it. Therefore, the first hidden layer in any network was equal to one half the number of values in the measurement or one half the number of processing elements in the input layer.

RESULTS AND DISCUSSION

Complete development of each network took less than 5 hr. The best performing network was network 6, which used log-magnitude inputs and three hidden layers. Because the log magnitude essentially removes phase information, it is interesting that this information is not necessary to make the predictions. Figures 5, 6, and 7 are plots from this network. Each figure shows ANN predictions for one of the output parameters as circles and the actual value for the parameter being predicted as a line. This correlation is performed for all 75 test measurements. Figure 5 shows the prediction for the parameter related to "good" coatings, Figure 6 shows the prediction for the parameter related to "poor" coatings, and Figure 7 shows the prediction for the parameter related to bad data.

The worst performing networks were those using only data at 10 Hz and above—networks numbered 13 through 18. This is reasonable because the most information on coating performance is contained at lower frequencies. Plots for network 17 are shown in Figures 8, 9, and 10. Scatter is fairly high, but even this network with no low frequency information was able to predict accurately most of the time. Some networks, like this one, misclassified a few measurements and assigned a "good" or "poor" coating value of slightly less than 1.0 for some good or poor coatings. These networks therefore appeared to make the predictions with less confidence than others, being right most of the time but occasionally misclassifying a measurement. Even the networks which had the poorest predictions still gave the proper prediction most of the time. The misclassifications of a few measurements by some networks might be a result of a specific piece of data in the test set. A detailed examination of the test data and comparison to specific measurements that were not accurately predicted was not performed because of time constraints, but it is intended that this will be performed as a part of any future work.

In general, as the number of hidden layers of a given network type increased up to the maximum tested (three), the deviations of the predictions from the actual values for that network decreased, resulting in less scatter and better predictions. This is illustrated by the prediction of bad data by networks 3 through 6, which differ only by the number of hidden layers, shown in Figures 11, 12, and 13 and 7, respectively.

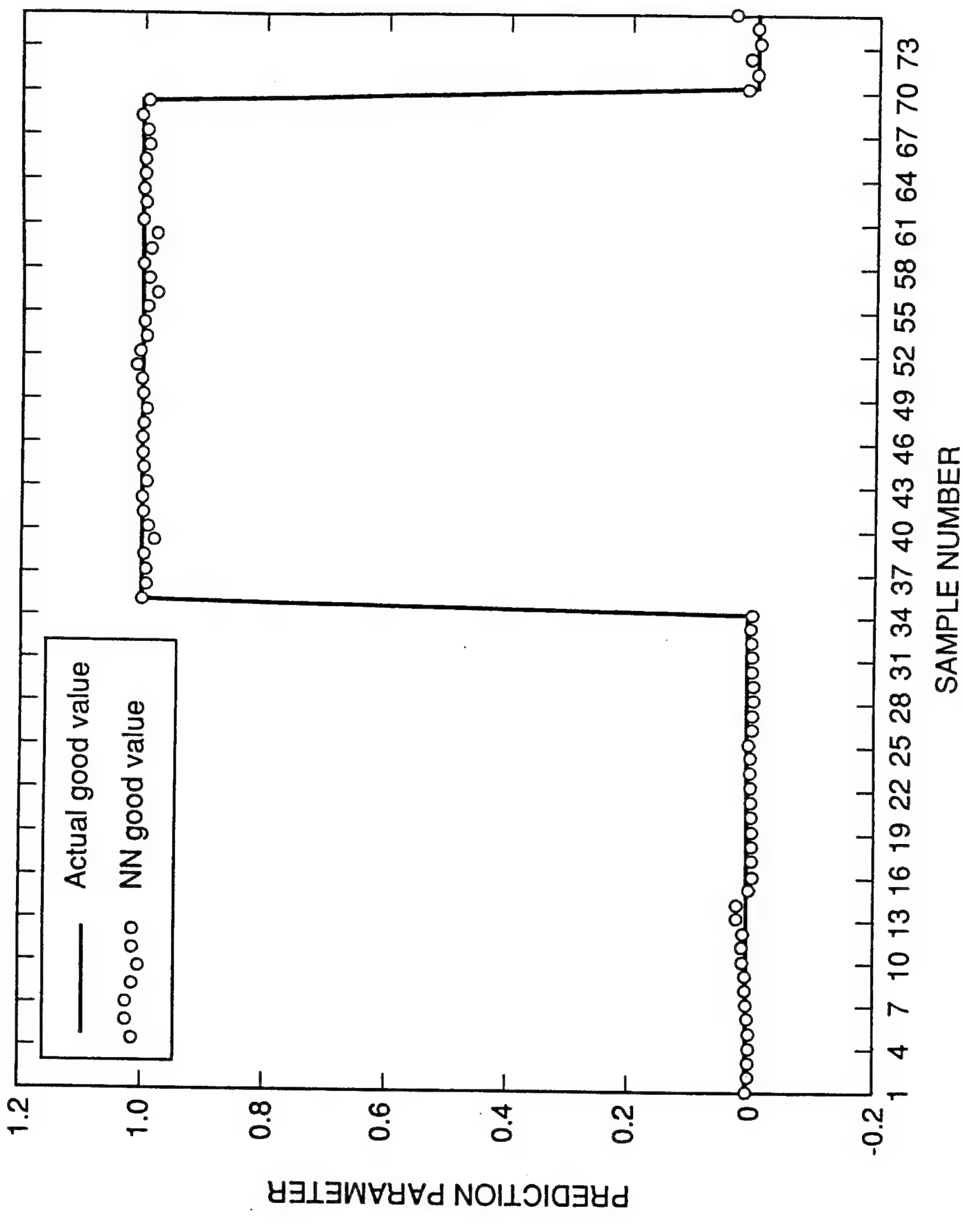

Figure 5. Network 6—"good" coating prediction.

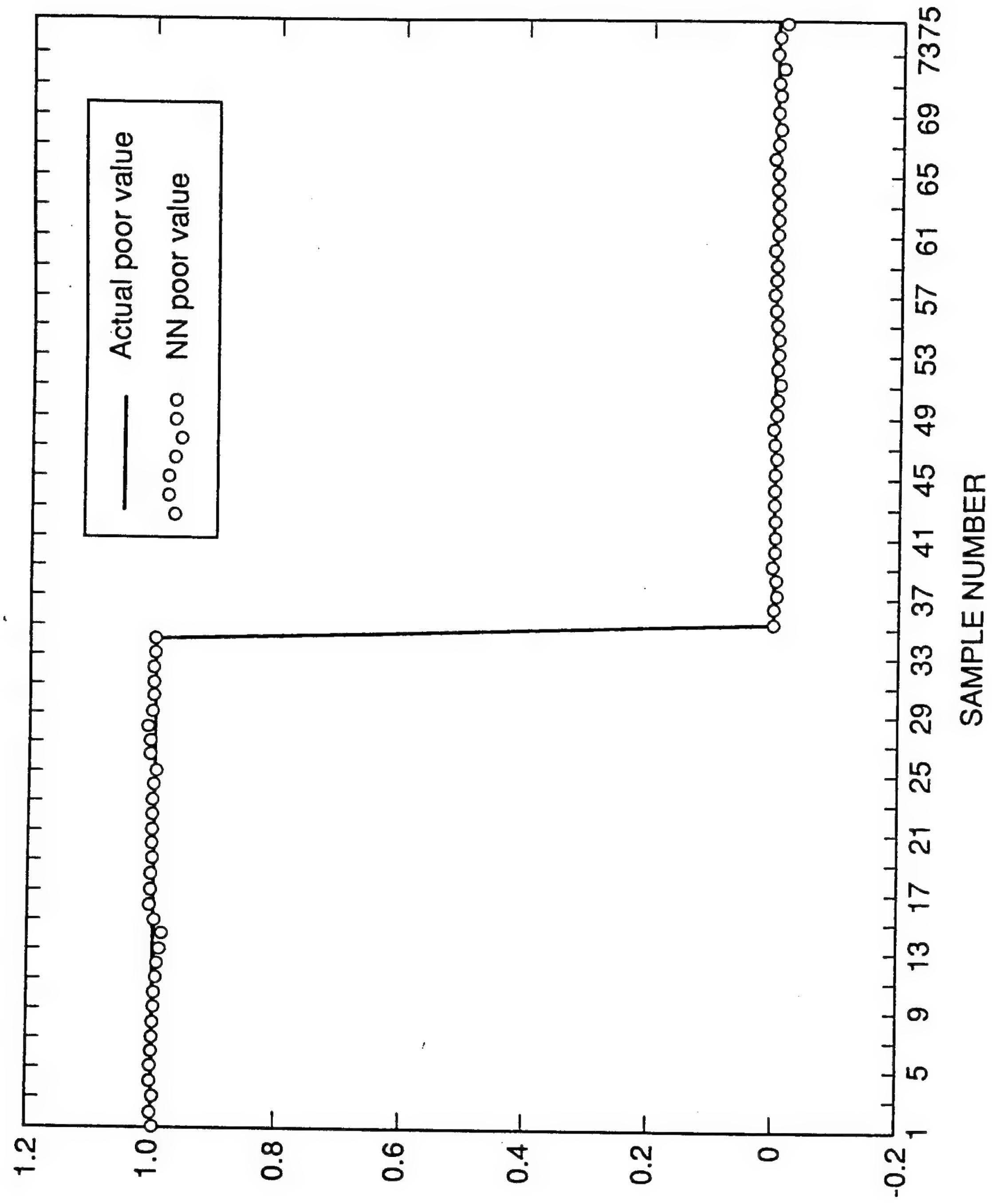

Figure 6. Network 6—“poor” coating prediction.

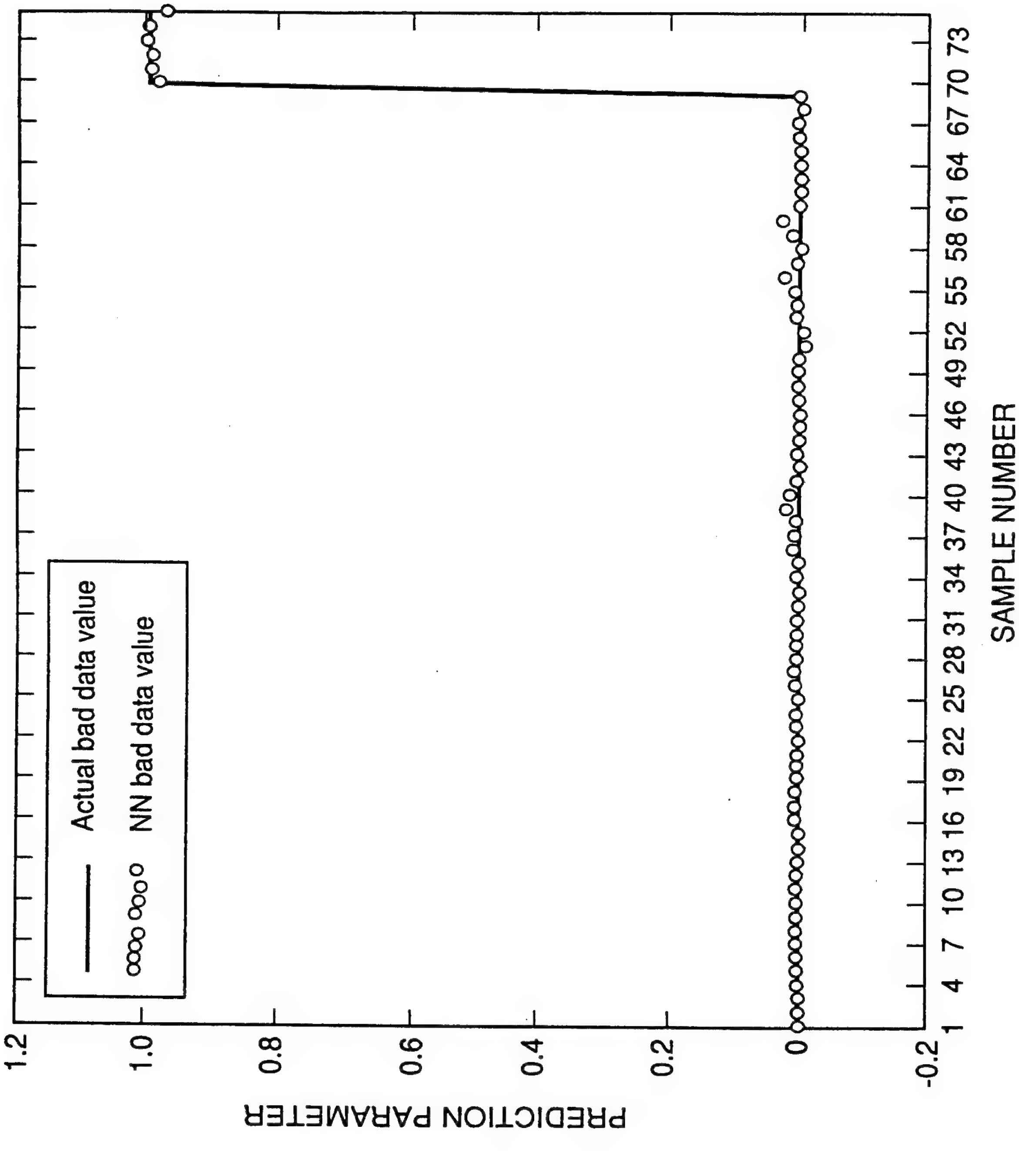

Figure 7. Network 6—"bad data" prediction.

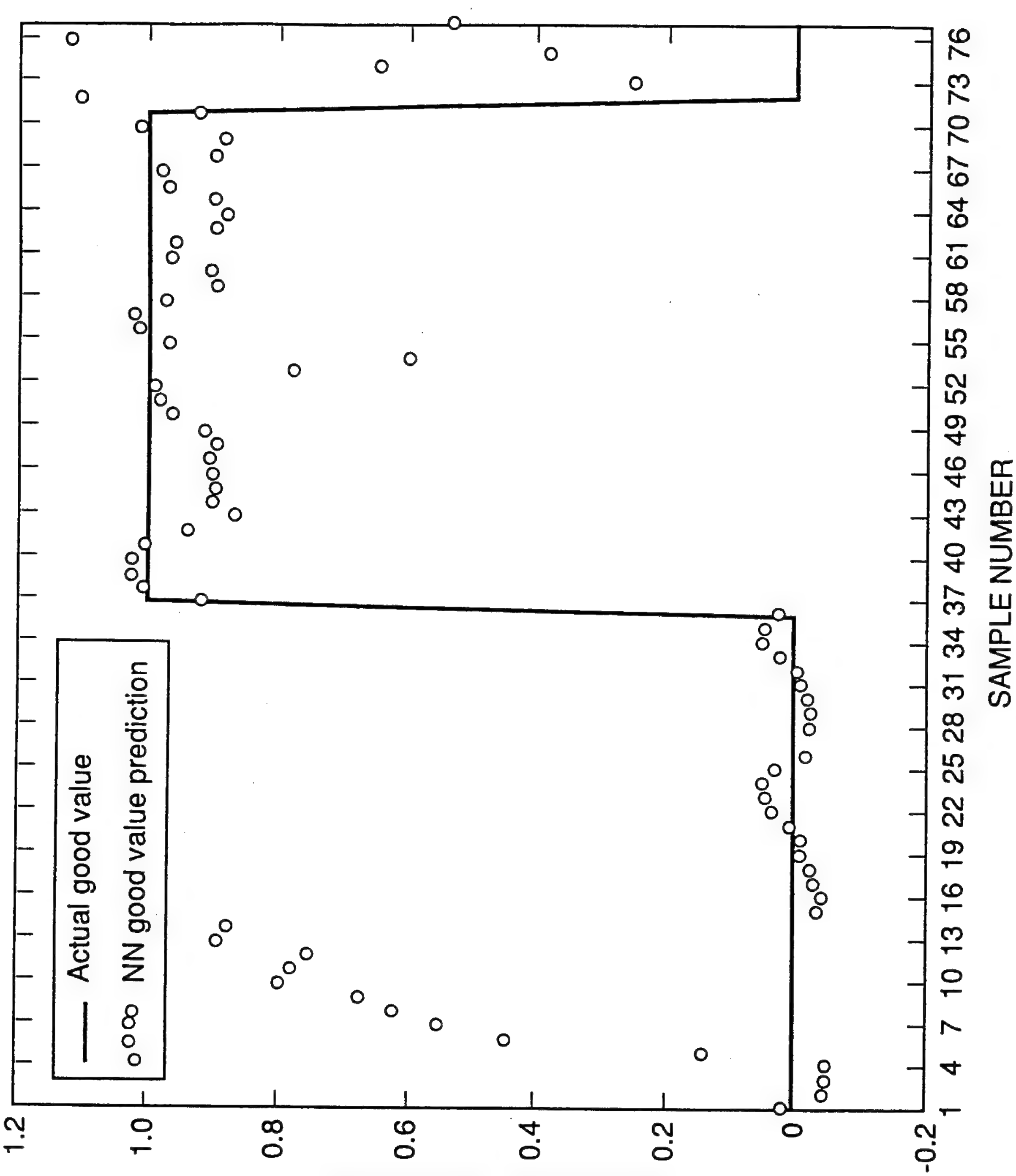

Figure 8. Network 17—"good" coating prediction.

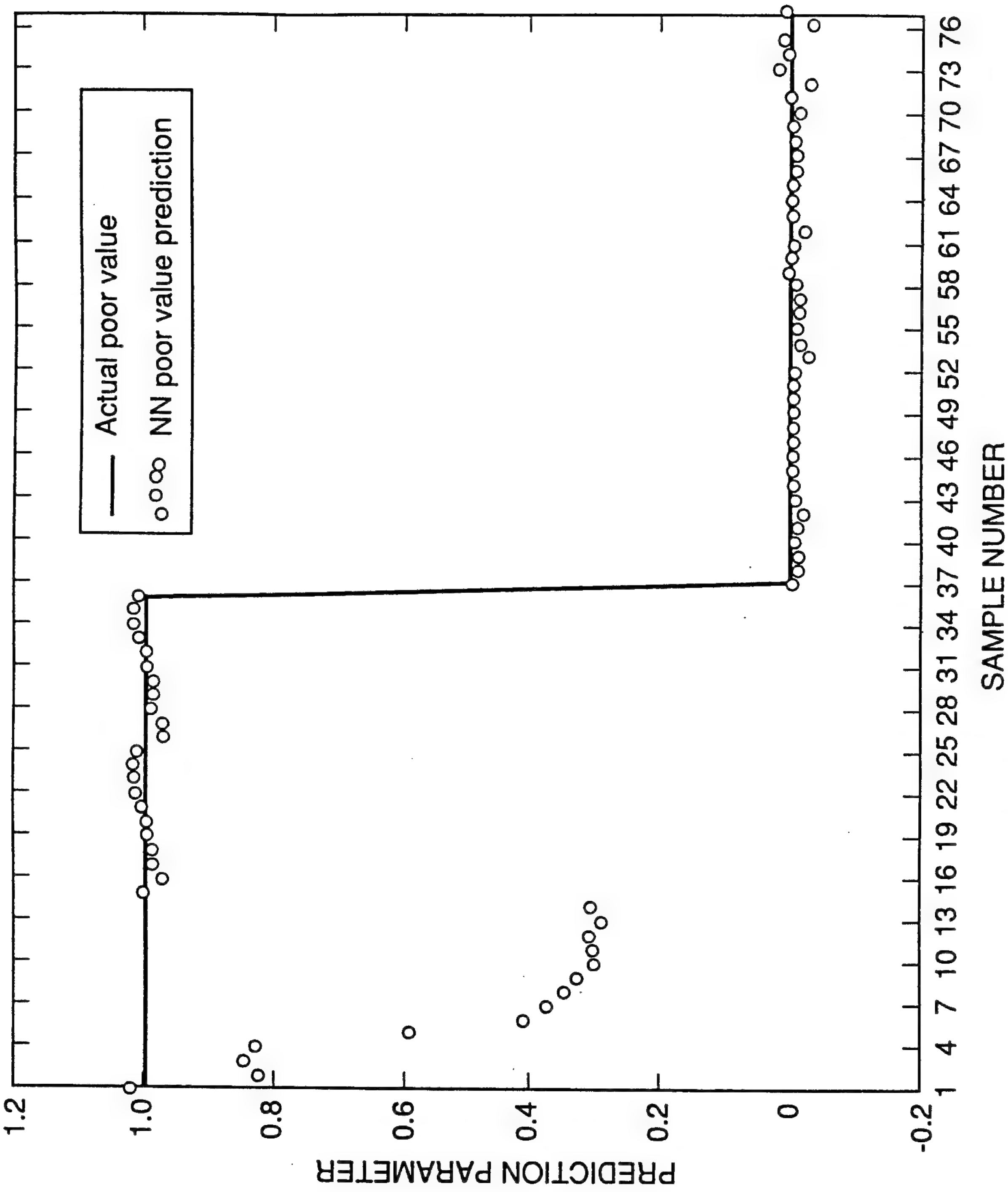

Figure 9. Network 17—"poor" coating prediction.

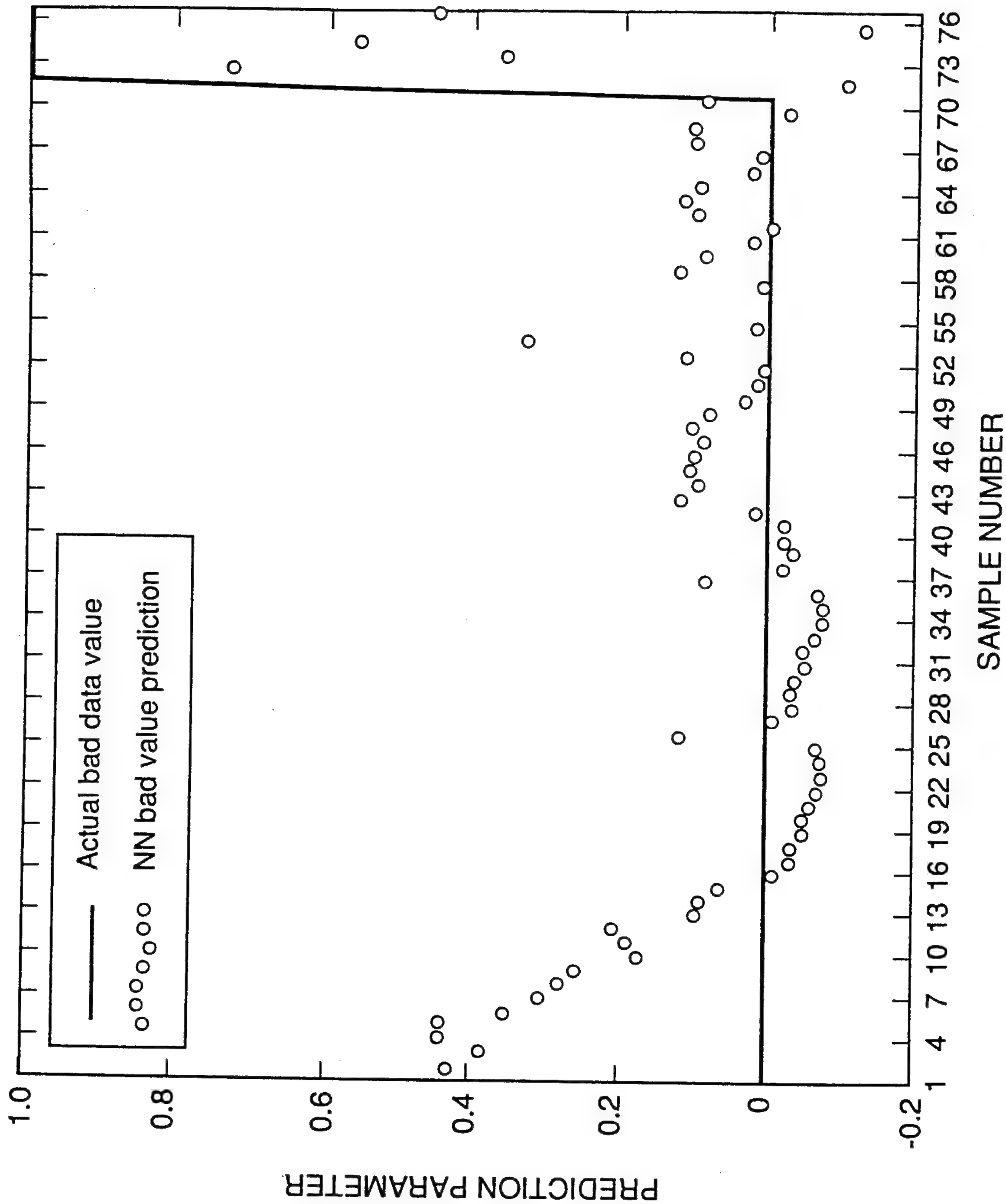

Figure 10. Network 17—"bad data" prediction.

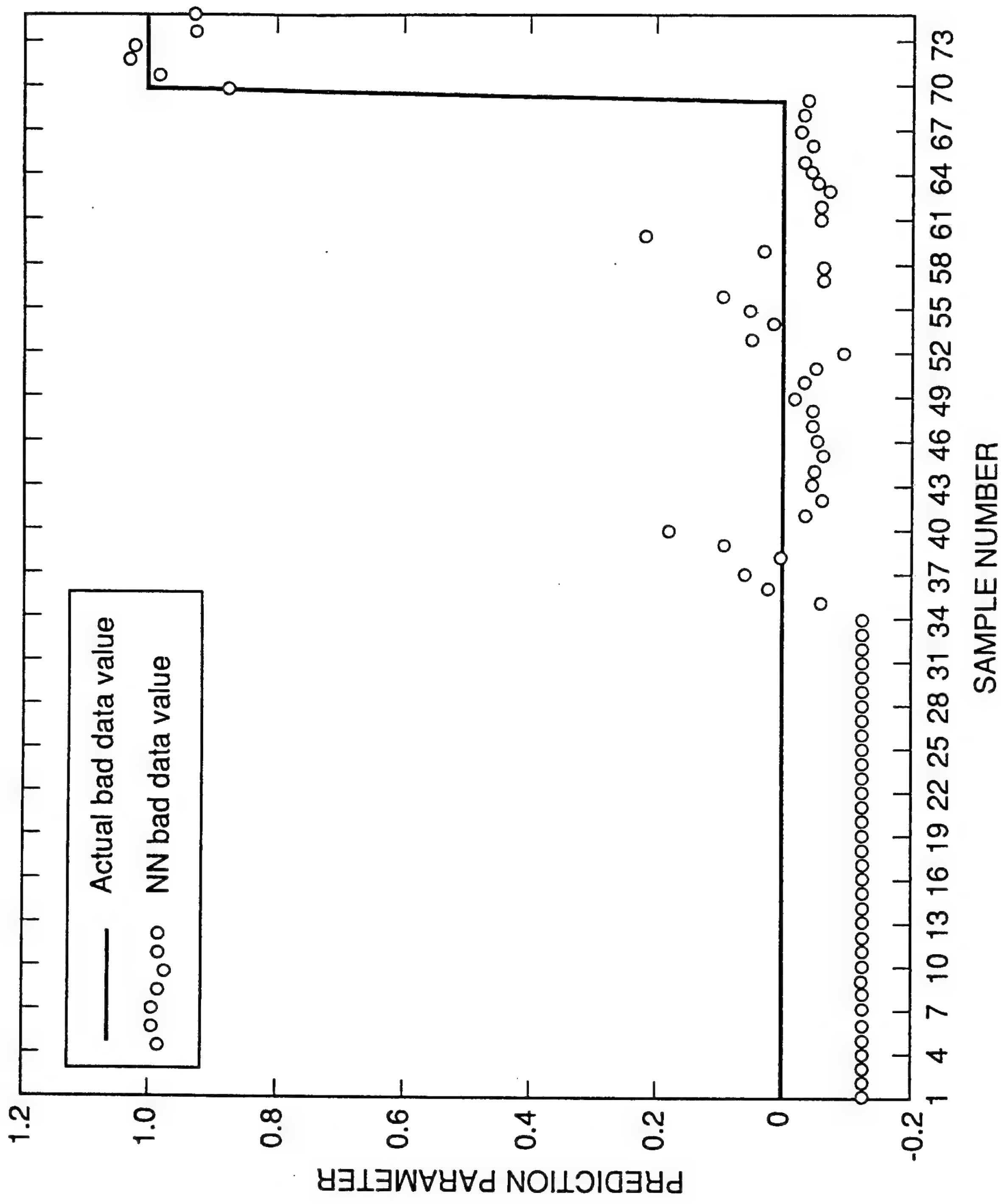

Figure 11. Network 3—"bad data" prediction.

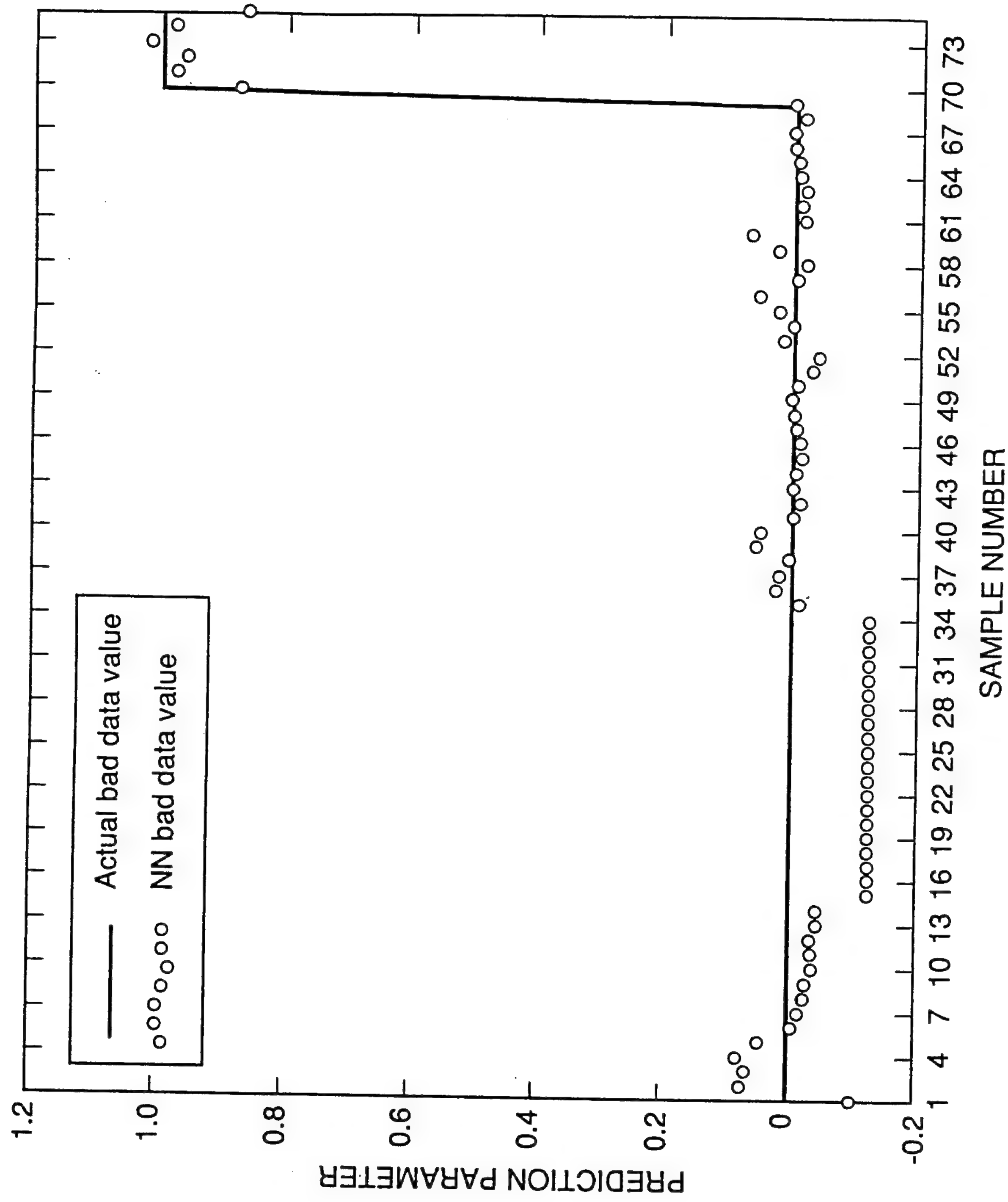

Figure 12. Network 4—"bad data" prediction.

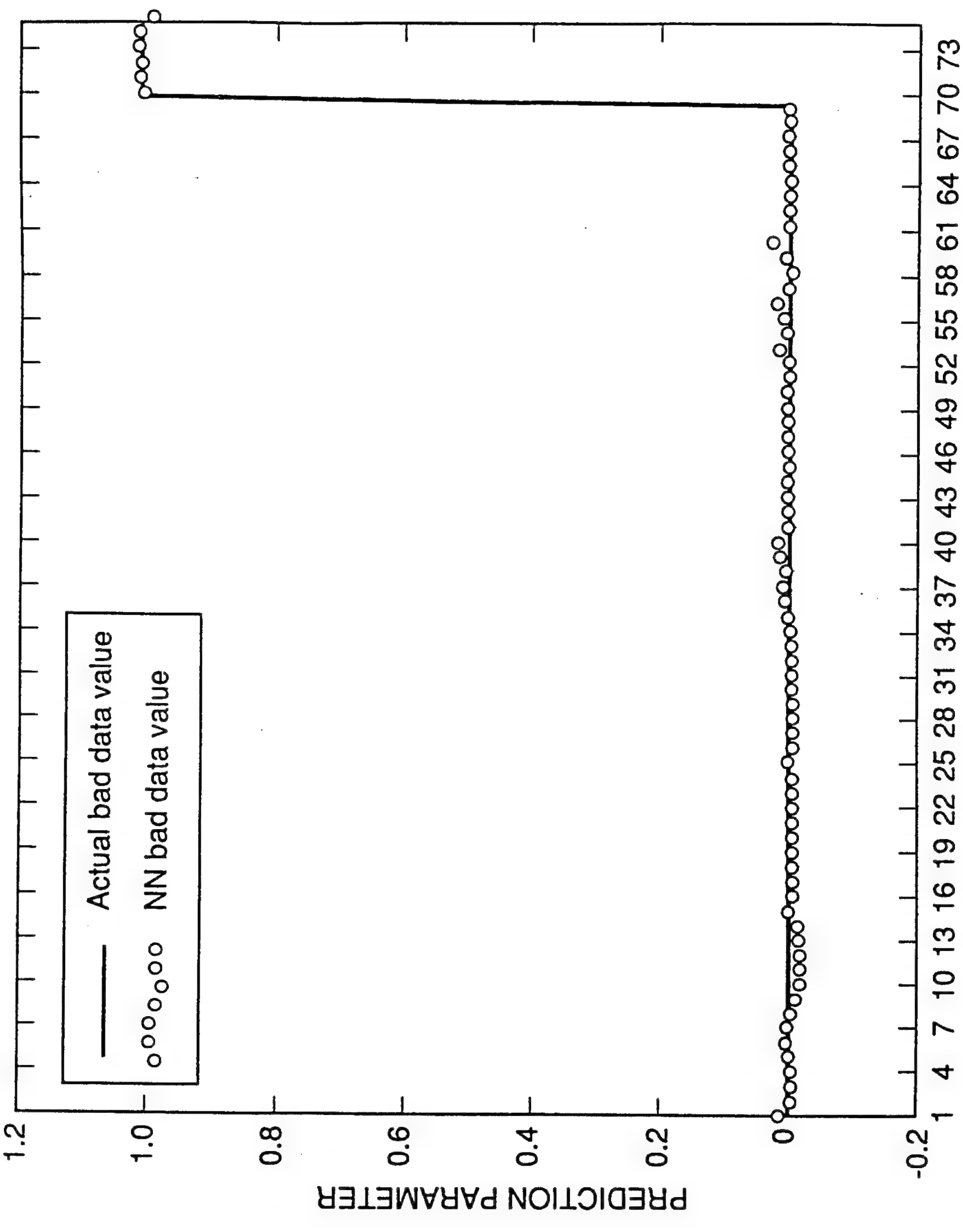

Figure 13. Network 5—"bad data" prediction.

A summary of the networks tested is included as Table 4.

Table 4. Networks studied.

Network Number	Input Scheme	Frequency Range	Input PEs	First Hidden PEs	Second Hidden PEs	Third Hidden PEs	Output Scheme	Output PEs
1	A	Full	142	50	0	0	A	3
2	A	Full	142	50	10	0	A	3
3	B	Full	71	0	0	0	A	3
4	B	Full	71	35	0	0	A	3
5	B	Full	71	35	15	0	A	3
6	B	Full	71	35	15	7	A	3
7	C	<1 Hz	52	20	0	0	A	3
8	C	<1 Hz	52	10	0	0	A	3
9	C	<1 Hz	52	0	0	0	A	3
10	D	<1 Hz	26	0	0	0	A	3
11	D	<1 Hz	26	10	0	0	A	3
12	D	<1 Hz	26	10	5	0	A	3
13	E	>10 Hz	60	20	0	0	A	3
14	E	>10 Hz	60	10	0	0	A	3
15	E	>10 Hz	60	0	0	0	A	3
16	F	>10 Hz	30	0	0	0	A	3
17	F	>10 Hz	30	20	0	0	A	3
18	F	>10 Hz	30	20	10	0	A	3
19	B	Full	71	0	0	0	B	3
20	B	Full	71	35	0	0	B	3
21	B	Full	71	35	15	0	B	3
22	B	Full	71	0	0	0	C	2
23	B	Full	71	35	0	0	C	2

The accuracy of the predictions of test data for all of the network runs is summarized in Table 5.

Table 5. Results.

Network Number	Input Scheme	Number of Hidden Layers	Output Scheme	Prediction Good Coating	Prediction Poor Coating	Prediction Bad Data
1	A	1	A	Fair	Fair	Poor
2	A	2	A	Poor	Good	Poor
3	B	0	A	Poor	Poor	Fair
4	B	1	A	Poor	Good	Good
5	B	2	A	Fair	Fair	Excellent
6	B	3	A	Excellent	Excellent	Excellent
7	C	1	A	Fair	Fair	Poor
8	C	1	A	Good	Fair	Poor
9	C	0	A	Fair	Fair	Poor
10	D	0	A	Fair	Fair	Fair
11	D	1	A	Good	Good	Good
12	D	2	A	Excellent	Excellent	Excellent
13	E	1	A	Poor	Excellent	Poor
14	E	1	A	Poor	Excellent	Poor
15	E	0	A	Fair	Fair	Poor
16	F	0	A	Fair	Fair	Poor
17	F	1	A	Poor	Poor	Poor
18	F	2	A	Fair	Poor	Poor
19	B	0	B	Poor	Poor	Fair
20	B	1	B	Excellent	Excellent	Good
21	B	2	B	Fair	Poor	Excellent
22	B	0	C	Fair	N/A	Fair
23	B	1	C	Fair	N/A	Poor

Using the linear transform of the real and imaginary impedances as input values made prediction of bad data difficult. Use of the logarithm of the total impedance as input resulted in good predictions, indicating that phase data are not required to get good predictions. Limiting the input data to frequencies of 1 Hz or below still resulted in good predictions, whereas using only data at 10 Hz and above resulted in poor predictions.

The best output scheme seemed to be where three output variables were used, indicating good coating, poor coating, or bad data, with each having values of zero to 1. Using a value of 0.5 for the good and poor coating values when the data were bad did not improve prediction accuracy and actually had a negative effect on accuracy. Using only two output variables, one indicating bad data with a range of zero to 1 and the other indicating good coating quality with a value of 1 or poor coating with a value of zero, resulted in poor predictions. The best network for predicting good and poor coating performance and for finding bad data was one using 71 input elements consisting of the log of the impedance magnitudes at each frequency; three hidden layers with 35, 15, and 7

elements, respectively; and three output elements indicating good coating, poor coating, and bad data.

CONCLUSIONS

Neural networks can be a very accurate tool for analyzing impedance data for painted steel. In a time span of less than 5 hr it was possible to develop a network capable of classifying E.I.S. data with an acceptable level of accuracy; minimal signal processing of the data was required to generate the input data for the network. However, proper construction of the network was necessary to ensure accuracy of the analysis and this construction would change if measurements over different frequency ranges or gradation schemes were used. All of the neural networks tested could determine with close to 100 percent accuracy if the coating was good or poor, and most networks tested could tell whether measurement problems produced bad data. Integration of a neural network classifier with an E.I.S. measurement system should be a simple, straightforward effort.

Additional work is underway to determine whether neural networks can be used for predicting values of parameters such as break-point frequencies, shelf impedances, and capacitances.

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13. ABSTRACT *(Maximum 200 words)*

Electrochemical impedance spectroscopy (E.I.S.) techniques can provide information about the condition of protective coatings on steel marine structures. Currently, an expert is required to interpret the data produced from an E.I.S.. measurement, classifying the coating as "good" or "poor" or identifying the data as "bad." This limits the use of E.I.S.. techniques to experienced operators. If the E.I.S.. technique is to be used for production by inexperienced operators, measurements must be classified automatically.

This investigation uses artificial neural networks (ANN) to develop an automated E.I.S. data classifier. ANNs were trained with a large database of measurements on known good or poor coatings, including some bad data. The ANNs were tested with E.I.S.. data not included in the training set. A variety of measurement signal processing schemes and network structures was evaluated. ANNs were developed which can accurately determine if the coating is good or poor and whether measurement problems produced bad data.

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